Thermodynamic Consistent Coupled Electrochemical-Mechanical Theory of Lithium-Ion Batteries

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The mechanical behaviour of lithium-ion batteries has a significant impact on their lifetime and their performance. The active particles of porous intercalation electrodes show significant volume changes due to lithium de-intercalation and intercalation. The reported volume changes of graphite are up to 10% of the nonlithiated structure and volume changes of silicon are greater than 300% compared to the de-lithated state [1]. Hence, the reference configuration of the porous solid materials is distorted and the porosity of the electrode is changed. However, as the porous electrode particles are surrounded by incompressible liquid electrolyte and the whole cell is included in a housing, the volume change of the electrodes is limited. This causes intercalation induced mechanical stress within the electrodes. The liquid electrolyte is exspelled from the porous structure. This displacement is, in turn, restricted by the housing and the counter electrode, which again induces pressure in the electrolyte. These strains and stresses may lead to battery failure mechanisms such as active particle fractures or electronic isolation of the active particles. In particular, this affects the electro-chemical kinetics of the cell. Therefore, the consideration of these effects in battery models is one of the major issues.

In this contribution, we develop a continuum model for the charge and discharge of lithium-ion electrochemical cells including mechanical effects of intercalation. The model is based on the theory of mixtures [2, 3] and extends the thermodynamic consistent mesoscopic battery model for porous electrodes and liquid electrolytes given in [4, 6, 5]. The constitutive models suppose incompressibility of both phases. In contrast to existing electro-mechanical theories [7, 8], the derived model includes not-only mechanical effects in the solid electrode materials but also the impact of the volume changes of the electrodes on the liquid electrolyte. Thus, for the first time, a coupled electrochemical and mechanical theory for whole electrochemical

cells is proposed.

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